THERMAL SHIFT AND BROADENING OF THE $^6A_{1g} \rightarrow {}^4A_1$, $^4E(^4G)$ PURE ELECTRONIC TRANSITION IN $RbMnF_3$*

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Received 20 February 1969

Temperature dependence of the shift and broadening of the 3975 Å line of $RbMnF_3$ is presented with comparison to the behavior predicted by simple spin wave theory due to Raman scattering of magnons.

The thermal shift and broadening of pure electronic transitions in magnetically ordered crystals has been the subject of several recent investigations [1-4]. The lines considered, for example in MnF_2, involved crystal field dependent states which made it difficult to estimate the relative importance of magnon and phonon processes. In addition, sizeable gaps exist in the spin wave spectrum of the compounds studied invalidating low temperature approximations which might be of help in determining the appropriate mechanisms.

The pervoskite antiferromagnet $RbMnF_3$ possesses a negligible anisotropy [5] and only nearest neighbor interactions are considered important [6], hence in most cases, simple spin wave theory is adequate to describe its low temperature magnetic behavior. $RbMnF_3$ is also known to remain cubic down to 2^oK [7] and to show a very small additional contraction due to magnetic ordering [8].

A sharp ($\Gamma_o \approx 2.5\ cm^{-1}$) magnetic dipole pure exciton line corresponding to a $^6A_{1g} \rightarrow {}^4A_1$, $^4E(^4G)$ transition is observed at 3975 Å in $RbMnF_3$ at 4.2^oK.

The excited state belongs to the same strong field orbital configuration as the ground state and is very insensitive to changes of Dq. Recent stress studies [9] show that the line shifts slightly to the red with increasing stress at a rate less than $3 \times 10^{-3}\ cm^{-1}/atm$.

We have measured the shift and width of this line using a crystal obtained from H. J. Guggenheim which allowed accurate data up to 40^o without multiple scanning. The line shifts $2.5\ cm^{-1}$ to the red and broadens $3.8\ cm^{-1}$ on warming to 40^o. Using published elasatic [10] and expansion [8] parameters along with the stress dependence of the line, an expansion shift of less than $0.15\ cm^{-1}$ is estimated in going from zero to 40^oK. Furthermore, the stress data suggest

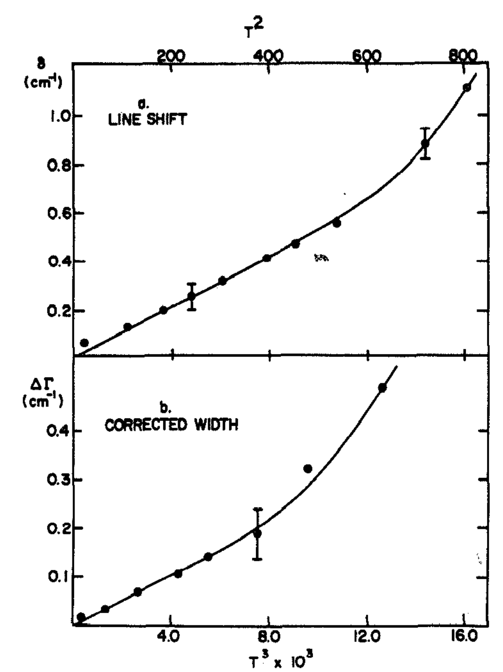

Fig. 1. (a) Line shift versus T^2 for the 3975 Å line of $RbMnF_3$. Shift was measured relative to the 3976.66 Å Cr line.
(b) Corrected width versus T^3. An intrinsic Lorentzian width equal to that measured at 1.5^oK has been used.

* Work supported by the National Science Foundation under Grant G. P. 7900 and the Wisconsin Alumni Research Foundation with funds administered by the Graduate Research Committee.
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that a blue shift due to expansion might be expected, and therefore the observed shift appears to be quite free of lattice effects. The effect of phonon scattering cannot be fully discounted, but would also seem to be small for this transition and temperature range.

An approximate calculation of the effect of Raman scattering of magnons on the shift and width of a pure electronic transition may be made by considering the optically excited ion essentially as an impurity interacting with its nearest neighbors via the Heisenberg exchange Hamiltonian. A first order perturbation calculation carried out in the limit of small k gives the shift in $RbMnF_3$ as

$$\delta\,(\mathrm{cm}^{-1}) = (2.35 \times 10^{-3})\,AT^2$$

where A is a parameter involving electronic matrix elements. In view of the small anisotropy gap, the small k approximation might be expected to be valid at very low temperatures in $RbMnF_3$. A similar calculation gives the temperature dependent width as

$$\Delta\Gamma(\mathrm{cm}^{-1}) = (2.55 \times 10^{-5})\,A^2T^3.$$

The measured shift and width are plotted against the predicted T^2 and T^3 dependence in fig. 1. The expected behavior is followed quite well up to about 23°, above which the approximations made might be expected to break down. The value of the parameter A required to fit the width data is within a factor of two of that appropriate for the shift results, and is of order unity as migh be expected for this transition. The thermal behavior of this line would seem to indicate that magnon scattering is the dominant shift and broadening mechanism at low temperatures in $RbMnF_3$. The deviation from simple spin wave behavior in $RbMnF_3$ reported in AFMR [11] and NMR [12] experiments is not noticeable below 20° in these measurements.

We wish to acknowledge correspondence with G. F. Imbusch, and to thank M. Y. Chen and D. S. Mc Clure for communicating the results of their stress studies to us.

References

1. W. M. Yen, G. F. Imbusch and D. L. Huber in Optical properties of ions in crystals, eds. H. M. Crosswhite and H. W. Moos (Interscience Publishers, N. Y., 1967) p. 301.
2. G. F. Imbusch, L. R. Walker, H. J. Guggenheim, W. M. Yen and D. L. Huber, preprint, unpublished.
3. J. Tylicki and W. M. Yen, Phys. Rev. 166 (1968) 488.
4. J. R. Chamberlain and J. T. Gourley, Phys. Lett. 26A (1968) 430.
5. P. H. Cole and W. J. Ince, Phys. Rev. 150 (1966) 377.
6. C. G. Windsor and R. W. H. Stevenson, Proc. Roy. Soc. 87 (1966) 501.
7. A. J. Heeger and D. T. Teaney, J. Appl. Phys. 35 (1964) 846.
8. D. T. Teaney, V. L. Moruzzi and B. E. Argyle, J. Appl. Phys. 37 (1966) 1122.
9. M. Y. Chen and D. S. Mc Clure, private communication.
10. D. E. Eastmen, Phys. Rev. 156 (1967) 645.
11. P. E. Seiden, Phys. Lett. 28A (1968) 239.
12. D. T. Teany, Bull. Am. Phys. Soc. Ser. II, 13 (1967) 164.

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